

THE LATTICE THERMAL CONDUCTIVITY OF SOME GOLD ALLOYS

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Summary

The electrical and thermal conductivities of the following gold alloys have been measured from 2 to 90 °K : Au-0.7 per cent. Pt, Au-1.6 per cent. Pt, Au-1.8 per cent. Pt, Au-0.65 per cent. Cr, and Au-6.2 per cent. Cr. Their lattice thermal conductivities have been deduced and the effects of annealing studied. It is found that in well-annealed dilute alloys, the lattice conductivity at liquid helium temperatures approaches a value consistent with theoretical predictions. It appears that increasing solute content causes an increasing dislocation density, which cannot be reduced substantially by annealing. This effect is more pronounced in the case of the chromium alloys. The lattice thermal conductivities at liquid oxygen temperatures compared with theory reveal numerical discrepancies, similar to those found in copper alloys.

I. INTRODUCTION

From the lattice component of the thermal conductivity of dilute alloys one can deduce the lattice conductivity of the pure parent metal and thus deduce the nature and strength of the electron-phonon interaction (see, for example, reviews by Klemens 1956, 1958). Thus, silver alloys were studied by Kemp *et al.* (1956) and copper alloys by several investigators, including White and Woods (1955), Kemp, Klemens, and Tainsh (1957), and Lomer and Rosenberg (1958). We have similarly investigated gold alloys ; a preliminary report of some of these measurements has already appeared (Birch, Kemp, and Klemens 1958).

The lattice thermal conductivity κ_g is limited by various processes, each of which contributes approximately additively to $W_g=1/\kappa_g$, that is,

$$W_g = W_E + W_D + W_U + W_P + W_I, \dots\dots\dots (1)$$

where W_E is the resistivity due to the scattering of phonons by conduction electrons, W_D by dislocations, W_P by point defects, W_I by static imperfections other than dislocations and point defects, and W_U is the resistivity due to three-phonon interactions.

Now W_E and W_D both vary as T^{-2} , and these resistivities are most important at liquid helium temperatures, while W_U and W_P , varying as T , are most important at liquid oxygen temperatures ; other imperfections have an intermediate temperature dependence and, if they affect κ_g significantly, do so at intermediate temperatures.

Most interest lies in the value of W_E , obtained from the value of $W_g T^2$ at liquid helium temperatures in well-annealed specimens. Cold-worked specimens have higher values of $(W_g T^2)_{He}$, no doubt due to an appreciable dislocation

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resistance W_D . However, there is also some doubt as to whether annealed specimens are really sufficiently free of dislocations for W_D to be negligible, and values of $(W_g T^2)_{\text{He}}$ observed on annealed specimens may only be upper bounds for W_E .

It has been commonly observed that $(W_g T^2)_{\text{He}}$ increases with solute content, and, as this increase occurs irrespective of whether alloying increases or decreases the electron concentration, and also occurs if the electron concentration is unchanged (gold in copper measured by Kemp, Klemens, and Tainsh 1957), one suspects that this increase may be due to a dislocation resistance, the number of dislocations retained after equivalent annealing treatment increasing with solute content.

Kemp, Klemens, and Tainsh found support for this view from a reduction of $(W_g T^2)_{\text{He}}$ of three copper-zinc alloys when increasing the annealing temperature. If the annealing temperature is critical, then it should be possible to achieve further reductions in dislocation density by annealing just below the melting point. This is extremely difficult in copper-zinc alloys because of the danger of zinc evaporation, but is quite feasible in gold-platinum alloys, which were available to us and seemed in any case suitable for deriving κ_g of pure gold by extrapolation, since gold-platinum forms a continuous solid solution. We have therefore measured the gold-platinum alloys not only after annealing at moderately high temperatures (700–750 °C), but also after annealing just below the melting point. The alloys selected contained about 1.8 and 0.7 per cent. Pt respectively, and a less complete study was made of a 1.6 per cent. Pt alloy. It was found that the 0.7 per cent. Pt alloy showed a value of $(W_g T^2)_{\text{He}}$ when annealed, which was only slightly higher than the theoretical value of $(W_E T^2)_{\text{He}}$; other alloys, however, had lower lattice conductivities owing to the presence of dislocations.

Since terminal values of W_D seem in general to decrease with solute content, it was hoped to obtain low dislocation densities by using a solute element of high specific resistivity, thus lowering the solute content. An alloy of 0.65 per cent. chromium was therefore measured, but W_D remained high.

This alloy had been prepared by diluting a commercially available Au-6.2 per cent. Cr alloy, and, since the latter alloy shows an anomalous electrical resistance change on annealing (Linde 1954), its lattice thermal conductivity was also measured to search for a corresponding anomalous change of lattice conductivity on annealing. No such effect was found, but this alloy showed extremely high values of W_g both before and after annealing.

In addition to studies at helium temperatures, there is also some interest in the value of W_g/T at liquid oxygen temperatures, as this gives information about the magnitude of W_U and about W_P due to a known concentration of solute atoms.

II. MEASUREMENTS

The measurements of thermal and electrical conductivity were carried out in the range 2–90 °K using differential gas thermometers and a galvanometer amplifier, with common points of attachment on to the specimen rod. The apparatus used has been described by White (1953) and by Kemp *et al.* (1956).

TABLE I
ELECTRICAL RESISTIVITIES AND LATTICE THERMAL CONDUCTIVITIES

Specimen	Condition	ρ_0 ($\mu\Omega$ cm)	$\rho_{90}-\rho_0$ ($\mu\Omega$ cm)	$\rho_{293}-\rho_0$ ($\mu\Omega$ cm)	$(\chi_g/T^2)_{He}$ (W cm $^{-1}$ deg $^{-3}$)	$(\chi_g/T)_{Ox}$ (W cm $^{-1}$ deg $^{-2}$)
Au-0.7% Pt 2.6 mm dia.	Drawn (80% red.)	0.857	0.61	2.25	5.8×10^{-4}	4.3
	Ann. 750 °C	0.816	0.63	2.27	2.6×10^{-3}	6.2
	Ann. 1050 °C	0.820	0.62	2.29	2.6×10^{-3}	6.2
Au-1.6% Pt 3 mm dia.	Ann. 700 °C	1.64	0.71	2.47	1.75×10^{-3}	—
	Ann. 1050 °C	2.04	0.66	2.28	1.8×10^{-3}	6.0
Au-1.8% Pt 3 mm dia.	Ann. 700 °C	1.85	0.66	2.40	1.5×10^{-3}	4.6
	Ann. 900 °C	1.83	0.72	2.46	1.6×10^{-3}	4.6
	Ann. 1082 °C	1.95	—	2.52	1.8×10^{-3}	4.6
Au-6.2% Cr 2 mm dia.	As drawn	27.3	3.9	5.8	1.07×10^{-4}	4
	Ann. 200 °C	28.8	3.5	4.7	1.43×10^{-4}	4
	Ann. 500 °C	27.9	3.4	4.9	2.2×10^{-4}	4
Au-0.65% Cr 3.2 mm dia.	As drawn	3.04	—	1.8	4.0×10^{-4}	—
	Ann. 500 °C	2.83	0.64	1.95	1.1×10^{-3}	7
	Ann. 1050 °C	2.96	—	—	1.2×10^{-3}	7
Pure gold	—	—	0.60	2.20	$2.9 \times 10^{-3*}$	35†

* Theoretical estimate (Klemens 1954).

† Theoretical estimate (Leibfried and Schloemann 1954; see also Klemens 1958).

The gold-platinum specimens were prepared by Messrs. Garrett, Davidson, and Matthey (Sydney), the approximate compositions being checked from values of the residual electrical resistivity, using data of Linde (1939; see also Gerritsen 1956) for the specific resistivity of platinum in gold (1.01 $\mu\Omega$ cm per atomic per cent.). In the case of Au-1.6 per cent. Pt the composition deviated appreciably from the expected composition (1 per cent. Pt), and this was confirmed subsequently by chemical analysis.* Since this discrepancy was not satisfactorily explained, we are inclined to give less weight to the results from this alloy. The specimens were received in the "as drawn" condition and were annealed *in vacuo* at the temperatures stated in Table 1 for 4 hr. The 0.7 per cent. Pt alloy was homogenized at 1050 °C and then drawn, reducing its cross section by 80 per cent.; in this case the three specimens were distinct, but in all other alloys annealing was cumulative.

The Au-6.2 per cent. Cr alloy (composition in atomic per cent.) was supplied by Messrs. Sigmund Cohn (New York); annealing was in an atmosphere of helium for 12 hr. It was later diluted about ten times with pure gold by Messrs. Garrett, Davidson, and Matthey to provide Au-0.65 per cent. Cr. The latter alloy was annealed *in vacuo* cumulatively for 4 hr.

* We are indebted to Mr. D. A. Sinclair, Branch Superintendent, Defence Standards Laboratory, Sydney, for this analysis.

III. RESULTS

Electrical resistivity data of the various specimens are summarized in Table 1. The thermal conductivities of some of the specimens are given as function of temperature in Figures 1 and 2.

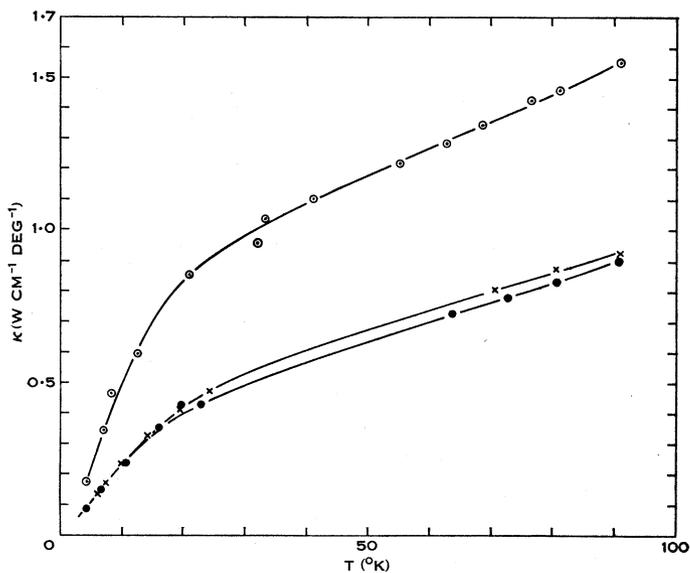


Fig. 1.—Thermal conductivity of gold-platinum alloys. Au-0.7% Pt annealed at 1050 °C ○; Au-1.6% Pt annealed at 1050 °C ●; Au-1.8% Pt annealed at 1082 °C ×.

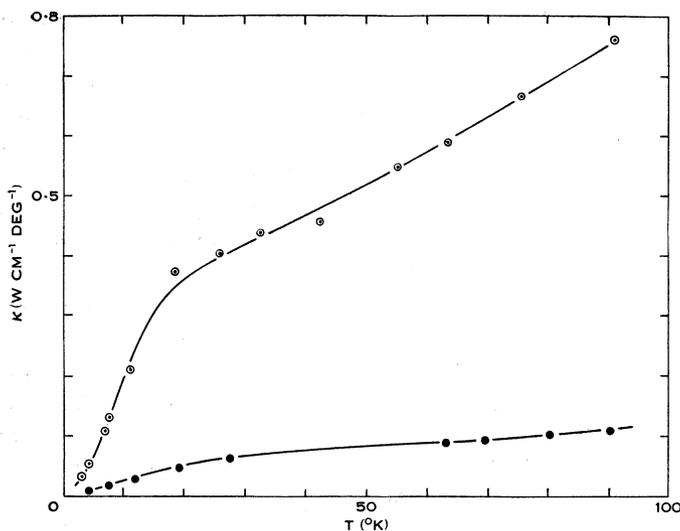


Fig. 2.—Thermal conductivity of gold-chromium alloys. Au-0.65% Cr annealed at 1050 °C ○; Au-6.2% Cr annealed at 500 °C ●.

The lattice thermal conductivity was deduced from measured values of $\kappa(T)$ and ρ_0 (residual electrical resistivity), as described, for example, by Kemp *et al.* (1956) and by Klemens (1958), using the relations

$$\kappa = \kappa_e + \kappa_g, \dots\dots\dots (2)$$

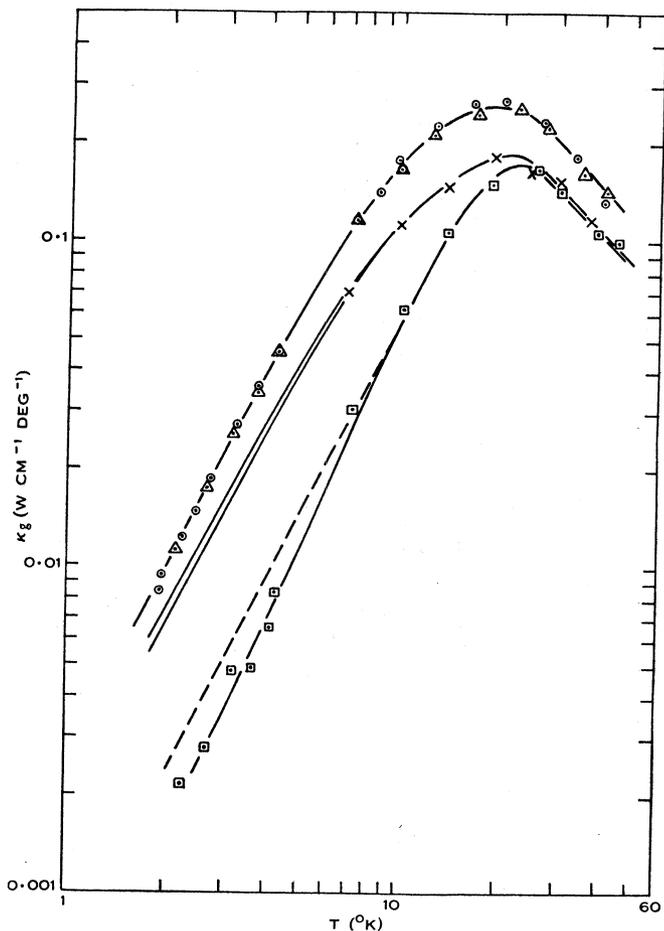


Fig. 3.—Lattice thermal conductivity of gold-platinum alloys. Au-0.7% Pt deformed \square , annealed at 750 °C Δ , annealed at 1050 °C \odot , dashed line: extrapolated T^2 variation; Au-1.8% Pt \times . For the latter alloy at low temperatures the upper curve is for specimen annealed at 1082 °C, the lower curve for that annealed at 700 °C; the coefficients of the T^2 dependence were determined from curves of κ/T against T , as described by Kemp *et al.* (1956).

where the electronic conductivity κ_e is given by

$$1/\kappa_e = W_i + \rho_0/LT, \dots\dots\dots (3)$$

$L = 2.45 \times 10^{-8} \text{ V}^2/\text{deg}^2$ being the Sommerfeld value of the Lorenz number and W_i the ideal electronic thermal resistivity. For W_i we took the values for pure

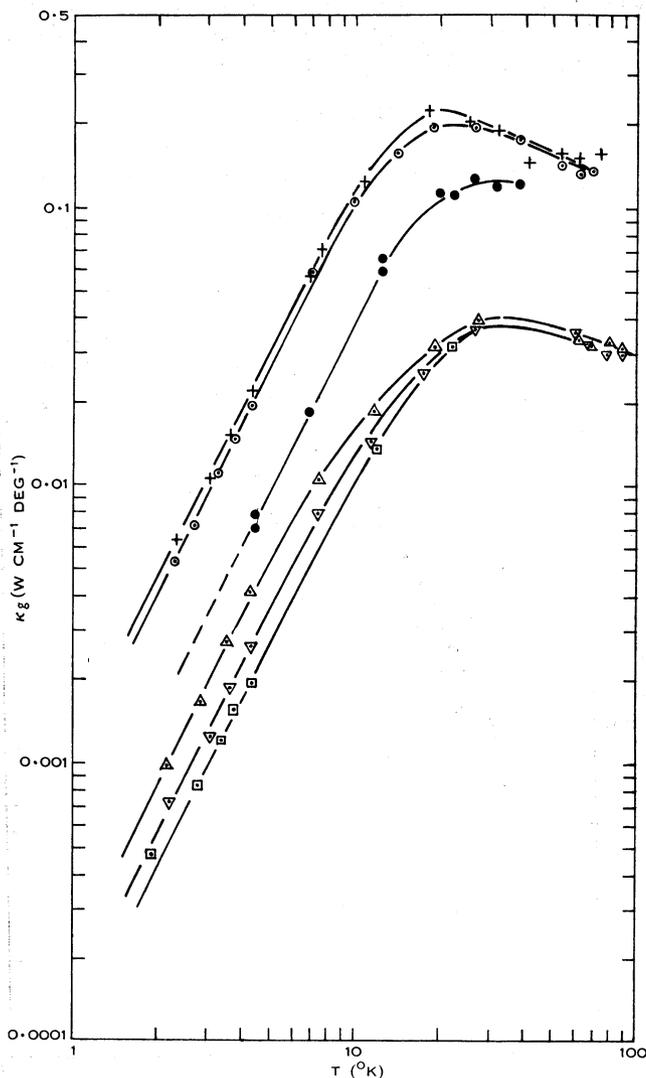


Fig. 4.—Lattice thermal conductivity of gold-chromium alloys. Au-0.65% Cr deformed ●, annealed at 500 °C ○, annealed at 1050 °C +; Au-6.2% Cr deformed □, annealed at 200 °C ▽, annealed at 500 °C △.

gold (White 1953), corrections due to the change of W_i with composition (Klemens 1959) being unimportant in the present alloys.

The values of κ_g for the various alloy specimens are displayed in Figures 3 and 4, except for the two Au-1.6 per cent. Pt specimens, the curves for which are practically coincident with that of Au-1.8 per cent. Pt annealed at 1082 °C. Values of κ_g/T^2 at helium temperatures and of κ_g/T at oxygen temperatures are also shown in Table 1.

IV. DISCUSSION

(a) *Low Temperature Results*

Consider the lattice conductivities at temperatures well below 20 °K. It is seen that $(\kappa_g/T^2)_{\text{He}}$ is least for the 6.2 per cent. Cr alloy, is higher for the 0.65 per cent. Cr alloy, higher still for the 1.8 and 1.6 per cent. Pt alloys, and highest for the annealed 0.7 per cent. platinum alloy. In general it seems sufficient to anneal at moderate temperatures (500–750 °C) to increase $(\kappa_g/T^2)_{\text{He}}$ substantially: annealing above 1000 °C produces relatively small changes only, and none in the 0.7 per cent. Pt alloys. Thus, while some reduction in dislocation density can be effected by annealing close to the melting point, the most important factor in determining $(\kappa_g/T^2)_{\text{He}}$ is the composition; chromium is much more effective in decreasing $(\kappa_g/T^2)_{\text{He}}$ than platinum.

The present results do not allow a clear distinction between the possibilities that the increase of $(W_g T^2)_{\text{He}}$ is due to a change in W_E or to an additional dislocation resistance. The insensitivity of the 0.7 per cent. Pt and, to a lesser extent, the 0.65 per cent. Cr and the 1.6 per cent. Pt alloys to further annealing at 1050 °C would indicate a change in W_E , but the results for the 1.8 per cent. Pt alloy, showing a relatively small change from 700 to 900 °C, but a larger change from 900 to 1082 °C, indicate a sluggish removal of dislocations, which requires very high annealing temperatures, and may well be incomplete.

Irrespective of why $(W_g T^2)_{\text{He}}$ increases with solute content, we obtain from the two annealed Au-0.7 per cent. Pt specimens an estimate of $(W_E T^2)_{\text{He}}$ which is probably not greatly in error. From the present results $(W_E T^2)_{\text{He}} > 0.38 \times 10^3 \text{ W}^{-1} \text{ cm deg}^3$. White, Woods, and Elford (1959) have recently obtained* a slightly lower value of 0.36×10^3 for a dilute gold-iron alloy (0.13 per cent. Fe). The theoretical value of $(W_E T^2)_{\text{He}}$ for gold is 0.35×10^3 , obtained by Klemens (1954) from the observed value of W_i/T^2 , assuming $\theta_D = 170 \text{ °K}$ and further assuming that the electrons interact equally strongly with transverse and with longitudinal phonons. On the other hand, the assumption of no interaction between electrons and transverse waves, but coupling between the polarization branches, leads to $(W_E T^2)_{\text{He}} = 7 \times 10^3 \text{ W}^{-1} \text{ cm deg}^3$. The present results, and those of White, Woods, and Elford, clearly favour the former coupling scheme, as proposed by Makinson (1938). Thus gold behaves similarly to copper and silver in this regard.

Since the Fermi surface of gold is most probably not spherical (Klemens 1956; Cohen and Heine 1958), the agreement between the observed values of $(W_g T^2)_{\text{He}}$ and the theoretical estimate of $(W_E T^2)_{\text{He}}$ confirms the theoretical prediction that the ratio W_E/W_i is not sensitive to small departures from sphericity.

If we assume that W_E is the same for all alloys and changes in $(W_g T^2)_{\text{He}}$ with composition are due to dislocations, we can deduce $(W_D T^2)_{\text{He}}$ for the various specimens and calculate the corresponding dislocation densities, using a relation

* We are indebted to Dr. G. K. White for communicating these results in advance of publication.

obtained by Klemens (1955) as subsequently modified by a numerical factor (Klemens 1958) :

$$W_D T^2 = 0.17 \frac{v h^2}{K^3} N b^2$$

$$= 3.9 \times 10^{-9} N \text{ W}^{-1} \text{ cm deg}^2, \quad \dots \dots \dots (4)$$

where N (in lines/cm²) is the dislocation density, v the sound velocity, h and K the Planck and Boltzmann constants, and b the Burgers vector. In Table 2 are given values of $(W_D T^2)_{\text{He}}$, obtained by assuming the theoretical value of 0.35×10^3 for $(W_E T^2)_{\text{He}}$, and also values of N calculated using (4).

TABLE 2
DISLOCATION DENSITIES

Specimen	Condition	$W_D T^2$ ($10^3 \text{ W}^{-1} \text{ cm deg}^2$)	N (10^{11} cm^{-2})
Au-0.7% Pt	Drawn (80% red.)	1.4	3.6
	Ann. 750 °C	0.04 (?)	0.1 (?)
	Ann. 1050 °C	0.04 (?)	0.1 (?)
Au-1.8% Pt	Ann. 700 °C	0.32	0.82
	Ann. 900 °C	0.28	0.72
	Ann. 1082 °C	0.21	0.54
Au-0.65% Cr	As drawn	2.1	5.4
	Ann. 500 °C	0.57	1.5
	Ann. 1050 °C	0.49	1.3
Au-6.2% Cr	As drawn	9.0	23
	Ann. 200 °C	6.6	17
	Ann. 500 °C	4.2	11

In attempting to explain the large number of dislocations in alloys remaining after annealing (comparable to the number introduced by deformation in the case of the chromium alloys) one naturally thinks of the movement of dislocations being impeded by the solute atoms. Either the solute atoms act as obstacles themselves or some other imperfections, associated with the solute atoms, act in this way. The action of impurities as obstacles has recently been demonstrated by Johnston and Gilman (1959) in the case of lithium fluoride. There are two effects: the impurities not only anchor the dislocations and tend to make them immobile, but, if the dislocations have broken away and are mobile, their velocities are decreased by a very large factor. It is not unlikely that solute atoms in gold play a similar role.

It is interesting to note that the two less dilute alloys (1.8 per cent. Pt, 6.2 per cent. Cr) have curves of α_x against T which differ in shape from those of the other two alloys, for they appear truncated around 10 °K. Furthermore, there is less change on annealing in that temperature region than at lower temperatures (none in the case of 1.8 per cent. Pt). The thermal resistivity

at these intermediate temperatures is not inconsistent with a thermal resistivity due to stacking faults, for stacking faults should contribute a resistivity $W_s \propto T^{-1}$ (Klemens 1957). This identification cannot be made with confidence, but, if it is provisionally adopted, the stacking fault probability can be calculated from the additional thermal resistivity. If a stacking fault is regarded as an extended dislocation, then the dislocation densities of Table 2 and the stacking fault probabilities can be used to obtain the average width of the stacking fault ribbon. This turns out to be about 40 Å in the 6.2 per cent. Cr alloy and 100 Å in the 1.8 per cent. Pt alloy. It is possible that the high values of the dislocation density in these alloys are a result of the additional inhibition of dislocation movement due to these stacking faults.

(b) *Results at Liquid Oxygen Temperatures*

At temperatures above about $\frac{1}{4}\theta_D$ (θ_D is the Debye temperature) theory indicates that $W_U \propto T$ (e.g. review by Klemens 1958), while $W_P \propto T$ below and up to θ_D . Thus in the absence of other imperfections W_g should vary as T above about 40 °K. In the case of the platinum alloys κ_g varies indeed as T^{-1} above about 30 °K up to about 50 °K—at higher temperatures κ_g can no longer be evaluated with confidence, but presumably this temperature dependence continues to higher temperatures. Values of $(\kappa_g T)_{0x}$ shown in Table 1 are values taken in the former temperature region. In the case of the chromium alloys, κ_g varies more slowly than T^{-1} at the highest temperatures at which κ_g could be determined. Values of $(\kappa_g T)_{0x}$ derived from κ_g at 90 °K and shown in Table 1 will thus in general underestimate κ_g in the true T^{-1} region, which is presumably attained at higher temperatures.

The difference in $(\kappa_g T)_{0x}$ between the 0.7 per cent. Pt and the 1.8 per cent. Pt alloy is presumably due to a point defect resistance $W_P \propto T$, and most likely originates from the scattering of phonons by the platinum atoms. This would make the resistivity due to 1 per cent. Pt atoms $W_P/T = 5.0 \times 10^{-2} \text{ W}^{-1} \text{ cm}$. Correcting the observed value of $(W_g/T)_{0x}$ of the 0.7 per cent. Pt alloy for the contribution due to W_P of the platinum atoms, we obtain $(W_U/T)_{0x} = 0.125$, that is, the intrinsic value of $(\kappa_g T)_{0x}$ is about 8.0 W cm^{-1} . A similar extrapolation of the chromium alloys to zero chromium content is much less certain, but gives the same result. White, Woods, and Elford (1959) also obtained a value of 8 W cm^{-1} for $(\kappa_g T)_{0x}$ of their Au-0.13 per cent. Fe alloy.

This may be compared with a theoretical value of $\kappa_g T = 35 \text{ W cm}^{-1}$ derived, with some approximations, by Leibfried and Schloemann (1954). The discrepancy of a factor 4 is slightly greater than that found in copper and silver alloys (see Klemens 1958).

The thermal point defect resistivity W_P of platinum in gold is considerably higher than one would have expected from their small difference in atomic mass. Theoretically (Klemens 1955), the point defect resistivity is given by

$$W_P/T = \frac{12\pi^2}{0.90} \frac{\alpha^3}{\hbar v^2} n S^2$$

$$= 21.7 n S^2 \text{ W}^{-1} \text{ cm}, \quad \dots \dots \dots (5)$$

where a^3 is the atomic volume, n the defect concentration per atom, and S^2 a numerical constant. If a point defect scatters only in virtue of its mass difference,

$$S^2 = (\Delta M/M)^2/12, \dots\dots\dots (6)$$

M and ΔM being the normal atomic mass and the deviation from it. For platinum in gold, $S^2 = 0.9 \times 10^{-5}$, while the present results indicate $S^2 = 0.23$. The scattering of phonons by the platinum atoms is thus due to such effects as the distortion of the lattice around the solute atoms, and the difference in the binding forces due to gold-platinum and gold-gold adjacencies. A similar situation prevails in regard to zinc in copper and cadmium in silver (Klemens 1958).

For chromium in gold we obtain from the results of the present paper $(W_p/T)_{\text{ox}} = 2.0 \times 10^{-2} \text{ W}^{-1} \text{ cm}$ per atomic per cent., or $S^2 = 0.09$. From the mass difference alone, by equation (6), S^2 should be 0.045, so that the chromium atoms scatter not only due to the mass difference, but also through misfit.

It is interesting to note that in the case of the 0.7 per cent. Pt alloy, but not in the case of the two chromium alloys, $(W_g/T)_{\text{ox}}$ is significantly larger in the deformed state than after annealing. This indicates the removal of point defects during annealing with a value of $nS^2 = 3.2 \times 10^{-3}$; but the electrical resistivity change during annealing (which may not be entirely due to point defects) is far too small to account for any point defect in concentrations of 10^{-2} to 10^{-3} per atom. The same difficulty was also found in the case of copper-zinc alloys (Kemp *et al.* 1957; Kemp, Klemens, and Tainsh 1959).

(c) Electrical Resistivity Changes

The change in ρ_0 of Au-0.7 per cent. Pt during annealing to 750 °C is small, but comparable with similar changes commonly observed in pure gold. All gold-platinum alloys showed an increase in ρ_0 on increasing the annealing temperature to 1050 °C or higher. This may be due to contamination or to the release of platinum atoms that had segregated. The latter hypothesis seems specially likely in the Au-1.6 per cent. Pt. alloy, partly because the increase in ρ_0 is too great to be accounted for by any likely contamination and partly because the solute atoms were probably not uniformly distributed, as indicated by the deviation of the apparent platinum content from the expected composition.

The Au-0.65 per cent. Cr alloy showed a similar increase after annealing at 1050 °C.

The Au-6.2 per cent. Cr alloy showed an increase of ρ_0 after annealing, similar to that previously found by Linde (1954) on annealing gold-chromium alloys of more than 2 per cent. Cr at 450–500 °C. The present results show that this increase occurs already when annealing at 200 °C; further annealing at 500 °C causes a small decrease, as in normally behaved metals.

One possible mechanism for the resistance increase on annealing is to suppose that some chromium atoms are segregated along the dislocations after deformation; the low temperature anneal causes them to disperse into the material, thus increasing the resistivity. On this basis the observed resistance increase would indicate that 5 per cent. of the chromium atoms were thus segregated,

i.e. about 0.003 per atom. On the other hand, a dislocation density of $2 \times 10^{12} \text{ cm}^{-2}$ (see Table 2) could accommodate only about 0.001 chromium atoms per atom, even if every dislocation were decorated by one atom per atomic plane. While a more concentrated decoration is not impossible, it appears unlikely that this is the mechanism of the electrical resistance increase.

V. ACKNOWLEDGMENTS

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